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CITATION:

Okamoto, K ...[et al]. Nonradiative recombination processes of carriers in InGaN/GaN probed by the microscopic transient lens spectroscopy. REVIEW OF SCIENTIFIC INSTRUMENTS 2003, 74(1): 575-577

ISSUE DATE:

2003-01

URL:

http://hdl.handle.net/2433/50160

REVIEW OF SCIENTIFIC INSTRUMENTS VOLUME 74, NUMBER 1 JANUARY 2003

Nonradiative recombination processes of carriers in InGaN/GaN probed by the microscopic transient lens spectroscopy

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(Presented on 25 June 2002)

Temporally and spatially resolved observations of the nonradiative recombination (NR) processes of carriers in low dislocated GaN and InGaN/GaN were successfully obtained by using microscopic transient lens spectroscopy. The heat generations and conductivities of NR processes were detected by the signal intensities and the time profiles. We found that the thermal conductivities were not so different at the seed region (threading dislocation density (TDD) $=1-2\times10^{9}$ cm^{-2}) and the wing region (TDD$=1-2\times10^{6}$ cm^{-2}) of air-bridged lateral epitaxially grown GaN and InGaN/GaN, but the amount of heat generated at the wing regions was much smaller than that at the seed regions. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1519666]

I. INTRODUCTION

GaN-based optical devices such as light-emitting diode and laser diode have very strong emissions in spite of high threading dislocation densities (TDD$=10^{8}-10^{10}$ cm^{-2}).[1] For a wider application of GaN-based optical devices, the development of the emission efficiency has been expected. A nonradiative recombination (NR) process of carrier in a material is one of the most important processes to control the optical property because a great numbers of carriers decay by the NR process at room temperature. It had been reported that the threading dislocations (TD) in GaN might act as the nonradiative recombination centers (NRC).[2] In order to reduce the TDD in GaN and InGaN, the growth technique has been developed rapidly. Recently, low dislocated GaN (TDD$=10^{6}$ cm^{-2}) have been achieved by using the epitaxial lateral over growth technique.[3] However, remarkable enhancement of emission efficiencies of low dislocated GaN and InGaN has so far not been found.[3–6] The actual correlation between the NR process and the TDD are still unknown because the direct observation of the NR process has been very difficult. Recently, we succeeded in the direct detection of the heat generation of the NR process probed by the transient grating (TG) technique.[7,8] In this article, we developed the microscopic transient lens (MTL) technique for the selective time-resolved measurement of the heat generation and conduction of the NR processes.

II. EXPERIMENTAL SETUP

A frequency-tripled beam of a Nd:YAG laser (355 nm) and a He-Ne laser (633 nm) were used as pump and probe beams, respectively. Both beams were focused at a sample by an objective lens ($\times100$). Spot sizes of both beams were about 3 μm in diameter. By the excitation with a Gaussian spatially distributed pump beam, refractive index (n) in the sample is also spatially modulated by the modulation of the carrier density change (δN) and the temperature change (δT). Such modulations act as transient lenses. Focus/defocus of the probe beam at the transient lens was detected by a photomultiplier tube with an optical fiber. All measurements were at room temperature (23 °C).

The samples used in this study were grown by metal-organic chemical vapor deposition. Recently, Kidoguchi *et al.* accomplished air-bridged lateral epitaxial growth (ABLEG).[9] ABLEG-GaN has two regions, one is a seed region having high TDD ($1-2\times10^{9}$ cm^{-2}) and another is a wing region having low TDD ($1-2\times10^{6}$ cm^{-2}). The sample structure of ABLEG-GaN was shown in Fig. 1. We used three samples; bulk-GaN, ABLEG-GaN, and InGaN (35 Å)/GaN(105 Å), a three-quantum wells (3QW) structure grown on ABLEG-GaN.

III. RESULTS AND DISCUSSION

The time profile of the MTL signal taken for the bulk-GaN was shown in Fig. 2. This signal has two components; the spikelike dip component just after photoexcitation and the slow decay component. The fast component and the slow component are due to the defocus and focus of the probe beam at the transient concave and convex lens effects in the sample, respectively. From $(\partial n/\partial N)>0$ and $(\partial n/\partial T)<0$, we assigned the fast and slow components to the modulations of the carrier density and the temperature, respectively. The signal intensity and decay of the fast component represents the created carrier density and the carrier recombination/diffusion, respectively. The signal intensity and decay of the slow component represent the amount of the generated heat by the NR process and the thermal conductivity of materials,

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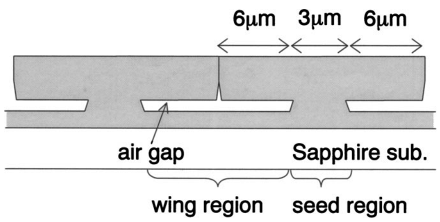

FIG. 1. Sample structure of the air-bridged lateral epitaxially grown (ABLEG)-GaN.

respectively. Time and spatial dynamics of δn and δT can be described by the rate equation including the diffusion/recombination of carriers and the generation/conduction of heat with cylindrical coordinates. By solving this rate equations, time and spatial dependence of δT can be written by

$$\delta T(r,t)=\frac{\tau_{\rm rad}}{\tau_{\rm rad}+\tau_{\rm nonrad}}\frac{QN_0}{\rho C_p}\left\{-\exp\left[-\frac{r^2}{w_0^2}\left(\frac{1}{\tau_{\rm rad}}+\frac{1}{\tau_{\rm nonrad}}\right)t\right]+\frac{w_0^2}{4D_{\rm th}t+w_0^2}\times\exp\left(-\frac{r^2}{4D_{\rm th}t+w_0^2}\right)\right\}, \tag{1}$$

where w_0 and r are the pump beam width and the distance from the beam center, respectively. N_0 is the initial density of carriers just after the excitation $(t=0)$. D and $D_{\rm th}$ are the diffusion coefficient of carriers and heat in GaN, respectively. Thermal conductivity (κ) of GaN can be obtained by $\kappa=D_{\rm th}\rho\ C_p$ with the density $(\rho=6.095\ {\rm g\,cm^{-3}})$ and the heat capacity $(C_p=9.745\ {\rm cal\,mol^{-1}\,K^{-1}})$. $\tau_{\rm rad}$ and $\tau_{\rm nonrad}$ are the radiative and nonradiative recombination lifetime, respectively. Q is the heat amount generated by the NR process. The optical pass of the probe beam at the transient lens given by the ABCD low of Gaussian beam. In this experimental condition, the time profile of the signal intensity of MTL $[S(t)]$ is proportional to $\delta T\ (t,r=0)$. Therefore, the decay component of the thermal signal can be described as

$$S(t)=\frac{I(t)-I(0)}{I(0)}\propto\frac{w_0^2}{4D_{\rm th}t+w_0^2}. \tag{2}$$

We could fit the slow decay component of Fig. 2 by using this equation and a value for $D_{\rm th}/w_0^2$ was obtained. To

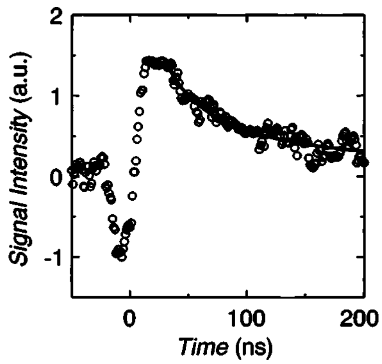

FIG. 2. Time profile of the microscopic transient lens spectroscopy taken for bulk GaN. Curved line is the fitting line by Eq. (2).

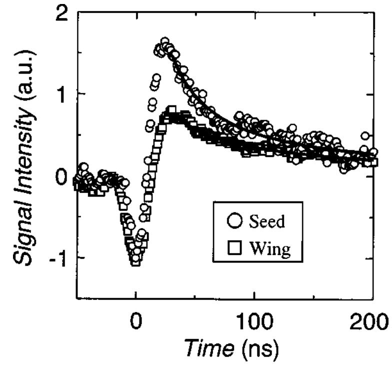

FIG. 3. Time profiles of the suite selective microscopic transient lens spectroscopy taken for ABLEG-GaN at seed region (○) and wing region (□). Curved lines are the fitting lines by Eq. (2).

obtain the $D_{\rm th}$ value, a value of w_0^2 is needed. We estimated $w_0=2.6\ \mu$m by using the $D_{\rm th}$ value for this sample obtained by the TG measurement $[D_{\rm th}=0.77\ {\rm cm^2\,s^{-1}}\ (\kappa=2.3\ {\rm W\,cm^{-1}\,K^{-1}})]$.[10] We can use this w_0 value for the measurement of ABLEG-GaN. The time profiles of the selective MTL signals at seed and wing regions in ABLEG-GaN were shown in Fig. 3. We fitted these decays by using Eq. (2) and obtained the $D_{\rm th}$ values at each region. The calculated κ obtained from $D_{\rm th}$ were $\kappa=2.2\ {\rm W\,cm^{-1}\,K^{-1}}$ at seed region and $\kappa=2.0\ {\rm W\,cm^{-1}\,K^{-1}}$ at wing region. κ values of each region were not so different and close to $\kappa=2.3\ {\rm W\,cm^{-1}\,K^{-1}}$ of bulk-GaN. κ of GaN had been measured as 1.3 ${\rm W\,cm^{-1}\,K^{-1}}$ by Sichel and Pankove in 1977.[11] Recently, Luo *et al.* reported that κ of GaN grown by the lateral epitaxial overgrowth (LEO) with TDD$=5\times10^6\ {\rm cm^{-2}}$ is 1.55 ${\rm W\,cm^{-1}K^{-1}}$ though κ of GaN with TDD$=10^{10}\ {\rm cm^{-2}}$ is 1.35 ${\rm W\,cm^{-1}K^{-1}}$ as probed by the scanning thermal microscope (STM).[12] In a similar way, Asnin *et al.* reported κ of GaN-LEO with TDD$>10^5\ {\rm cm^{-2}}$ as 1.7–1.8 ${\rm W\,cm^{-1}K^{-1}}$.[13] Florescu and co-workers reported that the κ values of GaN depend on the carrier concentration and thickness between 0.5 and 1.95 ${\rm W\,cm^{-1}K^{-1}}$.[14] They also developed the STM with spatial resolutions and reported that the κ values of GaN is 2.00 W cm$<\kappa<$2.10 W cm K on the partially overgrown regions.[15] Obtained κ values of bulk-GaN and ABLEG-GaN in this study are close to the maximum value which has ever been measured. This fact suggests that both our bulk-GaN and our ABLEG-GaN should have the highest crystallinity.

Although we could not find the difference of the κ values at each region, we found a remarkable difference of the thermal signal intensity at each regions. We could compare the thermal signal intensities by the NR processes of unit carrier densities at each region by normalization of the carrier signal intensities. It was found that the heat generation at the wing region was much smaller than that at the seed region. At the wing region, low TDD should reduce the NR processes of carriers. A similar result was obtained for the InGaN/GaN 3QW structure grown on ABLEG-GaN (Fig. 4). This fact suggests that the TD in GaN and InGaN must act as the NRC. For the first time, we have succeeded in detecting the heat generation from the TD and in clarifing the relationship between the NR process and TD in GaN and InGaN by

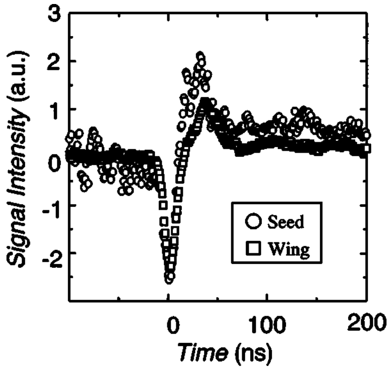

FIG. 4. Time profiles of the suite selective microscopic transient lens spectroscopy take for InGaN/GaN three quantum wells grown on ABLEG-GaN at seed region (○) and wing region (□).

the selective observation of the thermal signals. However, one question has remained: Why were optical properties at each region not different in spite of the three-order difference of TDD? From the measurement of the photoluminescence spectra, we knew that the emissions of these samples are dominantly attributed to free excitons and the internal efficiency of emission of each region is 0.6% at room temperature.[10] This fact means that the 99.4% of free excitons contribute to the heat generation. If internal emission efficiencies are decided only by the ratio between the radiative and nonradiative recombination processes of excitons, emission efficiency should be dramatically different at each region. We considered that the NR processes of excitons should not be a dominant path way of the thermalization in GaN and InGaN. We believe that another process must be the dominant thermalization path way, for example, slow thermalization processes after the thermal dissociation of excitons to electrons and carriers, or trapping into the deep energy level in the crystals. This is the possible reason that TD is not effective for the emission efficiency of GaN and InGaN/GaN though it acts as the NRC.

IV. CONCLUSION

Microscopic transient lens (MTL) spectroscopy is a powerful tool for the time and spatially resolved detection of the nonradiative recombination (NR) processes of carriers in GaN and InGaN/GaN. We found that the threading dislocations (TD) in GaN actually act as the nonradiative recombination centers (NRC) of created free excitons. However, TD should not be effective for the emission efficiency because another slow thermalization processes due to the thermal dissociation or the trapping in the deep level of excitons should be the dominant process for thermalization in GaN and InGaN/GaN.

ACKNOWLEDGMENTS

This work was partly supported by the Japan Society for the Promotion of Science (No. 12002454) and the Kyoto University-Venture Business Laboratory Project.

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